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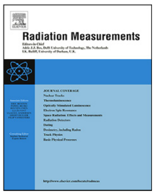

Fast neutron detectors and portal monitors based on solid-state heavy-oxide scintillators

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ABSTRACT

We have developed and fabricated large-sized scintillation blocks for detection of fast neutrons based on the heavy-oxide single crystals CWO, ZWO, BGO (diameter 40–50 mm, length 80–100 mm). These crystals demonstrate fast neutron detection efficiencies of not less than 50% without using an external plastic moderator. Based on these new detectors, we have created an experimental prototype of a portal monitor to detect mixed neutron/gamma radiation. The measured sensitivity for fast neutron detection was equal to or better than that of traditional ^{3}He gas discharge counters, with weight and size being lower by as much as an order of magnitude. It can be expected that this will also result in reduced production and operational costs. The interaction of fast neutrons with the heavy oxide scintillators was studied, and the respective contributions of the mechanisms of inelastic scattering of fast neutrons, resonance scattering of intermediate neutrons, and radiative capture of slow neutrons were determined. We have also developed an improved spectrometric "3-windows" method for separate detection of fast neutrons against the background of gamma-radiation.

1. Introduction

The use of ^{3}He gas counters in portal monitors for detection of illegal transport of special nuclear materials recently became problematic due to the limited supply of the helium-3 isotope (Kouzes et al., 2010, 2015). These counters are rather expensive; the current commercial price of helium-3 is about 3000 USD/liter, so typical large-area detector systems contain some tens of liters of helium-3 at a cost that may approach tens of thousands USD. Further, possible high-pressure helium-3 leakage (see, for example (Kouzes et al., 1911; Litvin et al., 2011),) can give rise to problems in their operation. Therefore, efforts to find alternative solid-state detectors for detection of fast neutrons have been actively pursued, see (Peerani et al., 2012; Lacy et al., 2011, 2013; Neutron detectors, 2017; Chandra et al., 2012; Arktis neutron detection, 2017; Alemberti et al., 2014; SaphyGate-GN, 2017; Fanchini, 2016; Paff et al., 2015; Cherepy et al., 2015; RPM, 2017) and others. For this purpose, one can use plates made of composite materials incorporating neutron-sensitive ^{6}Li or ^{10}B compounds, with possible additives of gadolinium or other neutron-capturing elements dispersed in an appropriate plastic or inorganic scintillator, e.g., Ag-activated zinc sulfide. Because of the low transparency of such dispersed materials, these detectors generally consist of thin active layers that are usually connected by different means to transparent plastic light guides (with or without spectrum-shifting dopants) to ensure that the scintillation light reaches the light-receiving device.

A certain success in the development of such solid-state detectors of thermal neutrons was reached using the composite material ^{6}LiF/ZnS(Ag) (see (RPM, 2017; Neutron detector, 2007; Dual Detector, 2017; Zaytsev et al., 2013; Marin et al., 2015) and others). At Los Alamos National Laboratory (Yanakiev et al., 2010), a multi-layer detector based on LiF foils was proposed, with its sensitivity comparable to the standard ^{3}He-detector HLNCC-II (18 helium-filled tubes pressurized to 4 bars). Several patents and publications describe various efforts to replace ^{3}He counters by

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solid-state neutron detectors. However, most of these technical solutions have an essential drawback. The alternative detectors, using neutron capture reactions by ^{6}Li or ^{10}B, detect sufficiently well only thermal neutrons (with detection probability above 50%), but not fast neutrons (less than 5% efficiency). To increase the detection efficiency of fast neutrons, one must incorporate an organic moderator of rather large thickness (and mass) before the detector to moderate the energies to thermal levels.

In our opinion, a very promising alternative approach to the creation of scintillation detectors for mixed neutron-gamma radiation is to rely on direct detection of fast neutrons within the scintillator material, with subsequent instrumental and/or software separation of the detected neutron fluxes and gamma radiation. Moreover, for the purpose of detection of illegally transported nuclear materials, such a combined n/γ detector could be more efficient because of the higher total sensitivity to the radiation emitted by such materials. Earlier, we developed a method for direct detection of fast neutrons using various inorganic scintillation single crystals (Ryzhikov et al., 2010, 2014, 2015, 2016; Grinyov et al., 2011) which is different from some other research to develop direct (without moderators) detection of fast neutrons using gas (Chandra et al., 2012), organic liquid (Paff et al., 2015) or plastic (Cherepy et al., 2015) scintillators. In this work, we report our development of portal monitors of fast neutrons on the basis of a "scintillator-PMT" type detector using special heavy-oxide scintillation single crystals. We carried out measurements of the efficiency and sensitivity of neutron detection with our new experimental installation, and developed an improved spectrometric method of "three windows" allowing discrimination of n/γ signals and selective detection of fast neutrons on the background of intense gamma-radiation.

2. Experimental apparatus

To measure the main parameters of scintillation detectors for fast neutrons, we created a new experimental installation. Its schematic design is shown in Fig. 1, and its general appearance – in Fig. 2. Using this installation, we carried out measurements of the efficiency and sensitivity of fast neutron detection with scintillators of different types and sizes.

At its basic length of 3 m, the installation has three operator-controlled (with the possibility of remote control) movable platforms for placement of detectors and radiation sources; two platforms are movable horizontally, and one platform – vertically. The positioning accuracy of each platform is 0.1 cm. Specially designed software allows measurements to be carried out in either a semi-automatic or an automatic regime.

For neutron sources, we used a ^{239}Pu-Be source with a neutron flux $0.95\text{x}10^{5}$ n/s (flux density at 100 cm from the source–0.76 n/s·cm^{2}) and a ^{252}Cf source with a neutron flux $2.35\text{x}\ 10^{6}$ n/s (flux density at 100 cm from the source–1.7 n/s·cm^{2}). The source was placed inside a lead sphere of 10 cm diameter with a 10 mm opening. The lead shield ensured discrimination of the accompanying gamma-radiation of the source. Additionally, the source could be coupled with a screen of gadolinium oxide to eliminate thermal neutrons. The neutron and gamma-radiation spectra of the sources are shown in Figs. 3–4.

A separate "scintillator-PMT" assembly or a complete fast neutron detector were placed into a light-protected housing and, if required, augmented by an additional lead screen with a collimator for protection from background gamma-radiation. The general scheme of the experiments is shown in Fig. 5. A Hamamatsu R1307 PMT with broad spectral sensitivity was used as the photoreceiver for these experiments.

For detection of mixed n/γ radiation, we used the heavy oxide single crystals CWO, ZWO, BGO produced by the Institute for Scintillation Materials, Kharkov, Ukraine, of up to 40–50 mm diameter and height up to 80–100 mm (see Fig. 6).

We carried out a set of measurements of neutron detection efficiency with the heavy oxide crystals CWO, ZWO, BGO of different sizes and volumes from 1 cm^{3} to 150 cm^{3}. Also, on our experimental apparatus we carried out measurements with other scintillator crystals of small sizes (1–4 cm^{3} volume) including single crystals of ZnSe(Te,O), YSO(Ce), NaI(Tl), CsI(Tl), ^{6}LiI(Eu), LuAG(Ce), LYSO(Ce), GSO(Ce) to determine how the neutron detection efficiency depended upon the scintillator type and its effective atomic number.

Using large-sized ZWO and BGO scintillators, we prepared for

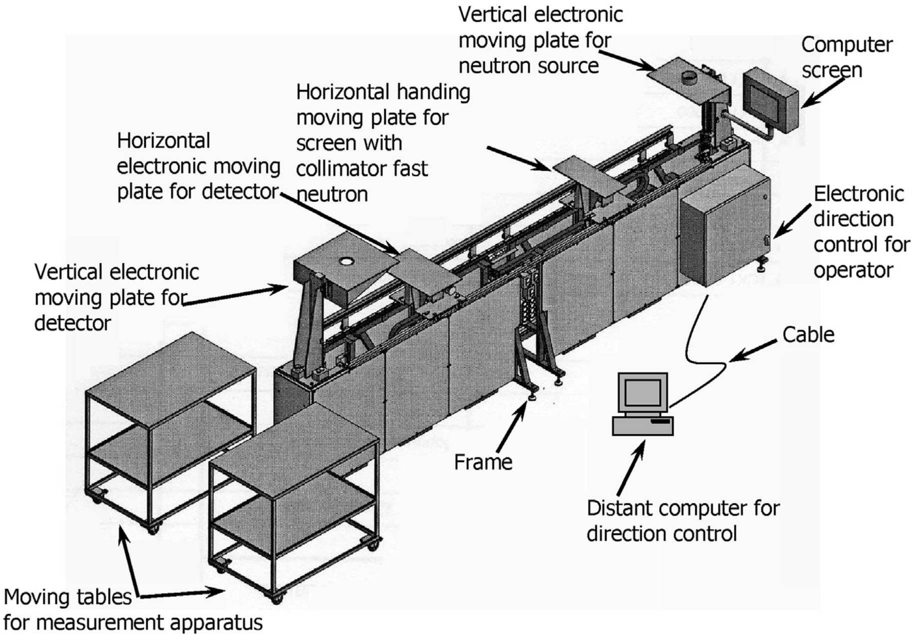

Fig. 1. Scheme of the experimental setup. Length of hardware system–3 m. Height–1.5 m. Position accuracy of electronic-controlled plate–0.1 cm.